

PATENT SPECIFICATION

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(54) IMPROVEMENTS IN OR RELATING TO SYNTHESIS APPARATUS

(71) We, CHEMIE LINZ AKTIEN-GESELLSCHAFT, (formerly Österreichische Stickstoffwerke, Aktiengesellschaft) an Austrian Body Corporate of St. Peter 224, Linz/Donau, Austria, do hereby declare the invention, for which we pray that a Patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to an improved synthesis apparatus and more particularly to an apparatus for guiding a gas in a catalytic high pressure synthesis plant, for example for the synthesis of ammonia.

In catalytic ammonia high pressure synthesis reactors with the catalyst arranged in several successive layers, it is known that the gas flows through the first catalyst layers by heat exchange, within the reactor, with the fresh gas which enters, the synthesis gas flowing axially through all layers (see Austrian Patent Specifications Nos. 216,533, 225,722 and 215,436).

Installations are also known in which the gas flows through the first catalyst layers axially, but through the long terminal catalyst layer radially from the interior outwards (British Patent Specification No. 1204634), and here again, *inter alia*, heat exchange, within the reactor, with the fresh gas occurs between the catalyst layers. The advantage over furnaces through which the gas only flows radially, and in which the gas flows alternately from the outside inwards and from the interior outwards, is that it avoids difficulties in forming during the reduction of the oxide catalyst for ammonia synthesis, which arise if the gas is guided radially from the exterior inwards.

The advantage over furnaces through which the gas only flows axially was considered to be the lower pressure loss, which manifests itself as a troublesome factor especially in the long terminal catalyst layer, whilst the pressure loss in the relatively low catalyst layers

at the input was regarded to be so slight that there was considered to be no advantage in guiding the gas radially in these catalyst layers. On the contrary, it was to be feared that in the catalyst layers at the input, where the reaction is particularly intense, losses in yield would occur, especially when using a catalyst of small particle size, as a result of local overheating, especially in the zones of the catalyst layers which are in the vicinity of the axis.

Surprisingly, it has now been found that in furnaces where heat exchange takes place within the reactor between the catalyst layers, the synthesis of ammonia may be improved by at least 10 to 13%, relative to the customary furnaces through which the gas flows axially, if the gas flows from the interior outwards in a radial direction in all catalyst layers and preferably if a catalyst of a particle size of less than 3 mm is employed.

The present invention provides an advantageous apparatus containing a device which is particularly suitable for guiding the gas. Thus, according to the invention there is provided a catalytic high pressure synthesis apparatus containing a device for guiding a gas there-through, for example for the synthesis of ammonia, which apparatus comprises a high pressure vessel with an insert sleeve, at least two superposed catalyst layers located above a main heat exchanger, and two or more further heat exchangers located between the catalyst layers for heat exchange of reacted gas with the fresh synthesis gas which enters the high pressure vessel, optionally via the main heat exchanger, each catalyst layer being located in an annular space between two cylindrical perforated metal sheets which are concentric to a central device for guiding the gas through the pressure vessel and of which the inner perforated metal sheet forms, with the central gas guiding device, an inner annular space serving for the flow of gas from the central guiding device to the catalyst layers and the outer perforated metal sheet

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forms an annular space with the insert sleeve which serves to lead the gas from the catalyst layers to the main heat exchanger, the said perforated metal sheets being closed off by a container lid and a container base, the container lids in each case possessing an annular passage which is near the axis and opens into the inner annular space, and the container bases in each case having a passage near the insert sleeve and connected to the outer annular space.

The invention will be more particularly described with reference to the accompanying drawing which illustrates a preferred embodiment of the apparatus comprising three catalyst layers.

In the illustrated embodiment fresh synthesis gas enters through an inlet 1 into a high pressure container 2 having a furnace insert mantle 3. The fresh gas entering at 1 flows downwards through the furnace, via an annular space 4 between the insert sleeve and the high pressure container, from where it passes into the main heat exchanger 5. Catalyst layers 6 are located between concentric annular metal sheets 10 and 11 and are in each case delimited by container lids 8 and container bases 9. In the container lids 8, near the central gas guiding devices 13, are located gas inlets 14, whilst gas passages 15 located near the furnace insert mantle 3 are to be found in the container bases 9. An inner annular space 16 feeds the gas into the catalyst layer whilst the annular space 17 serves to lead the gas away from the catalyst layer. Intermediate heat exchangers 7 are located between the catalyst layers, and 18 is the gas outlet from the furnace. Concentric annular metal sheets 12 are welded onto the container lids 8, and these dip into the loosely poured catalyst and prevent a gas short-circuit as the catalyst settles. In this furnace construction, the central gas guiding device 13 is a central tube at the level of the lower catalyst layer. After leaving the first intermediate heat exchanger 7 it consists of a central tube around which are located two concentric passages of annular cross-section. The gas passes through the outer annular passage into the intermediate heat exchanger above it, and from there, after having its direction changed by 180°, it enters the central tube in which is located the burner 19. The gas passes from the central tube into the upper catalyst layer and is then returned to the level of the upper intermediate heat exchanger from whence it passes through the second catalyst layer and is then returned to the level of the first intermediate heat exchanger.

The equipment according to the invention may also be provided with feed devices which permit introducing cold fresh gas into the tubular interspace of the intermediate heat exchangers 7, in order to permit fine regula-

tion of the temperature in these heat exchangers 7.

The fresh gas which has passed through the annular space between the high pressure container and the furnace insert mantle enters the pipe interspace of the main heat exchanger 5, where it is heated by the synthesis gas which is leaving. It is passed through the central gas guiding devices 13 to the upper end of the furnace, and on its path it passes through the two intermediate heat exchangers 7 and is heated further. From the upper end it passes via the gas passage 14 of the uppermost catalyst layer 6 into the inner annular space 16 within this catalyst layer, from where it flows through the catalyst layer in a radial direction and passes via the annular space 17 and the gas passage 15 to the intermediate heat exchanger, where it is cooled. After flowing through all catalyst layers in the same manner, the gas leaves the furnace via the main heat exchanger 5 and the gas outlet 18.

The following Example illustrates the present invention and the manner in which it may be performed.

Example

Using an apparatus for the synthesis of ammonia as shown in the drawing, 75,000 N m³/hour of a synthesis gas mixture are passed radially from the interior outwards through three catalyst layers, at a pressure of 310 atmospheres gauge. Before entering the furnace the gas has the following composition:

67.5% by volume of hydrogen,
22.5% by volume of nitrogen,
8.0% by volume of inert gases, and
2.0% by volume of ammonia.

The entire catalyst volume is 2.2 m³.

The catalyst has been manufactured from a mixture of iron oxide corresponding to the composition Fe³O₄ with 0.5% by weight of potassium oxide, 2.8 to 3% by weight of aluminium oxide, 3.2 to 3.4% by weight of calcium oxide and 1% by weight of magnesium oxide, by reduction with a mixture of hydrogen and nitrogen in the furnace. It has an average particle size of 2 mm. The ammonia synthesis is carried out at 310 atmospheres and at a temperature of 400 to 500°C. On leaving the furnace, the gas contains 17.81% by volume of ammonia.

WHAT WE CLAIM IS:—

1. A catalytic high pressure synthesis apparatus containing a device for guiding a gas therethrough, which apparatus comprises a high pressure vessel with an insert sleeve, at least two superposed catalyst layers located above a main heat exchanger, and two or more further heat exchangers located between the catalyst layers for heat exchange of reacted gas with the fresh synthesis gas which enters

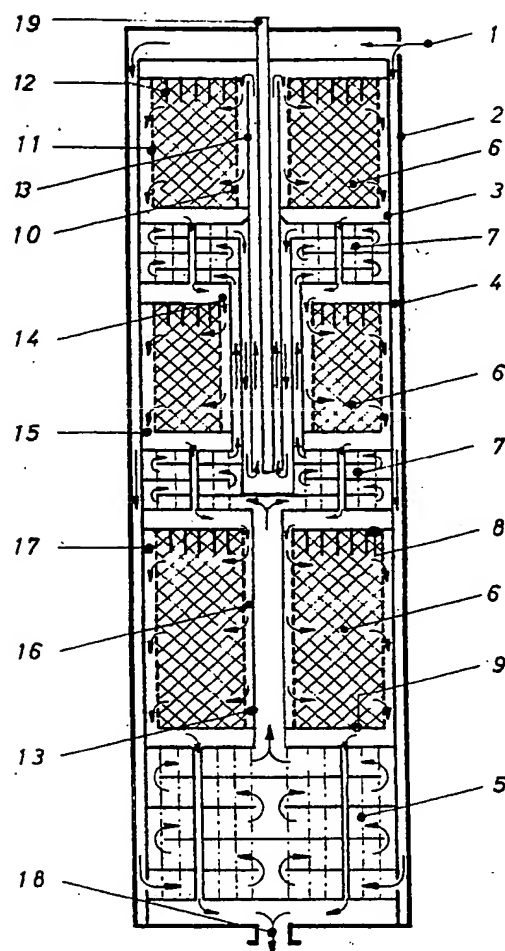
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- the high pressure vessel, each catalyst layer being located in an annular space between two cylindrical perforated metal sheets which are concentric to a central device for guiding the gas through the high pressure vessel and of which the inner perforated metal sheet forms, with the central gas guiding device, an inner annular space serving for the flow of gas from the guiding device to the catalyst layers and the outer perforated metal sheet forms an outer annular space with the insert sleeve which serves to lead the gas from the catalyst layers to the main heat exchanger, the said perforated metal sheets being closed off by a container lid and a container base, the container lids in each case possessing an annular passage which is near the axis and opens into the inner annular space, and the container bases in each case having a passage near the insert sleeve and connected to the outer annular space. 20
2. An apparatus according to claim 1, in which the inlet for synthesis gas is via the main heat exchanger. 25
3. An apparatus according to claim 1 or 2 which is adapted for the synthesis of ammonia. 30
4. A catalytic high pressure synthesis apparatus substantially as hereinbefore described with reference to the accompanying drawing.

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